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The dependence of the bubble density for an ionizing particle on the temperature and pressure in a bubble chamber

Part II. Comparison between theories and experimental results

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With 3 figures in the text

SUMMARY

The bubble nucleating mechanism in the bubble chamber is discussed in view of some of the existing experimental evidence for a description of the process, either by the occurrence of free electrical charges on the bubbles as required by the electrostatic theory or a localized heating of small regions in the liquid as given by the thermal theory. Calculations of the temperature dependence of operating conditions to be expected when either theory is correct have been made with the use of somewhat simplified models. The calculated temperature dependences have also been compared with those experimentally obtained by measurements of the bubble density as a function of the pressure drop and the temperature in the liquid for electrons of 18 MeV mean energy. The liquids used are propane, Freon-12 and sulphurhexafluoride. The experimental results for these liquids are in very good agreement with the behaviour of the bubble chamber as estimated from the thermal theory.

Introduction

The internal of a pure liquid is surprisingly stable against spontaneous boiling even when the liquid is strongly superheated because bubbles, smaller than a certain size, the critical size, will collapse as a consequence of the additional pressure on the bubbles arising from the surface tension. The bubble chamber is based on the fact that ionizing particles can initiate boiling under such conditions where there is only a small chance for the boiling to occur spontaneously. For practical as well as theoretical reasons the initial step of the boiling is of considerable interest. As yet no complete theory has been worked out which can explain in detail the phenomena observed in the behaviour of bubble chambers. The problem of bubble nucleating seems complicated and unknown enough to resist purely theoretical attacks but some progress can be made when allowing certain simplifications. The justification for simplified models is the agreement between the predicted and the observed operating conditions.

Theories on nucleation

Electrostatic theory

In his earlier papers on the bubble chamber Glaser [1–3] proposed that the nucleating of bubbles along the path of an ionizing particle passing a superheated liquid was analogous to the formation of drops in supersaturated vapours in the cloud chamber. The ionizing particles were thought to produce clusters of electrical charges with the same sign in localized volumes of the liquid. The electrostatic repulsion between the charges was then considered responsible for the ruptures in the liquid, which within a very short time appeared as macroscopic bubbles, provided appropriate conditions on the degree of superheat and temperature were fulfilled. If this electrostatic theory is a correct description of the initial step in a bubble chamber it should be able to predict the conditions of temperature and degree of superheat under which primary submicroscopic bubbles will grow to visible size.

The electrostatic theory has, no doubt, been successful in the sense that for several liquids it has been able to provide at least approximate estimates of the degrees of superheat which are required to make the liquids sensitive to ionizing radiation. Unfortunately, the calculations based on the model are strongly dependent on the number of free charges, assumed to be present on the bubble surfaces. The net charge cannot be measured by independent methods but can be obtained only by formulas derived from the theory. From experimental data available on a number of organic liquids at the temperatures and degrees of superheat usually employed in bubble chambers, the number of charges is found to be of the order of 6 when the ionizing particles are singly charged.

In spite of the fact that the electrostatic theory has had remarkable success in predicting operating conditions, there are several arguments which can be used against it. One is that it seems uncertain whether the electrical charges which are required by the model appear sufficiently often with the same sign in such small volumes of the liquid as is necessary [4]. The primary interactions between an ionizing particle and the atoms in the liquid are certainly too few to explain the observed bubble density. It has been found, however, that the bubble density along the path of an ionizing particle is inversely proportional to the square of the velocity of the particle [5, 6]. Thus, it varies in the same way as the number of δ-rays produced by the particle. This fact indicates that the majority of bubbles observed are produced in those regions of the liquid where the δ-electrons have stopped. Since the effective ranges of low energy electrons can be very short indeed because of the large scattering, which makes the electron movement more or less diffusive, it may very well happen that in strongly localized regions of the liquid quite a large ionization can occur, resulting in the production of many ion pairs. Part of these may then group in such a way that the net charge necessary for the production of bubbles of at least the critical size will be present in a sufficiently large number of regions in the liquid.

Another argument against the electrostatic theory which has been put forward is that ions, if initially present in the liquid with the concentration necessary, do not live for a time long enough for a bubble to grow to its critical size.

However, the time constant for recombination of ions with such a concentration that must be assumed here seems very uncertain. The estimated values are between 10^{-7} and 10^{-15} sec [7], while the time for the growth of a bubble to critical radius possibly can be as small as about 10^{-10} sec [8]. Because of the large uncertainty in the time constant for recombination it seems safer not to rule out the electrostatic theory on the argument given above.

Further, Glaser [4] has reported that while the electrostatic theory can possibly explain the initiating of boiling in a bubble chamber liquid (an exception is related below) caused by singly charged particles, an unreasonable large number of charges—all with the same sign—must be assumed to exist on the bubbles when α-particles are used as the ionizing agent.

Another discrepancy with the electrostatic theory was found by Glaser *et al.* [9] when liquid xenon was tried as a bubble chamber fluid. No distinguishable tracks could be obtained in xenon until about 2 % of ethylene had been added.

Xenon is a good scintillator and the effect of the ethylene is to quench the scintillations. The explanation is thought to be that xenon, being monatomic, has no rotational or vibrational degrees of freedom which can take care of the energy from excited ions or atoms by collisions of the second kind before these excited ions or atoms radiate their energy. By adding some ethylene, which has several degrees of freedom, most of the primary energy should rapidly be converted into kinetic energy and soon appear in the form of heat in the liquid.

Thermal theory

The experience which was gained from the experiments with the α-particles and the behaviour of xenon made it necessary to consider more seriously another possible nucleating mechanism. From the theoretical predictions by Fürth [10] based on the "hole" theory of liquids it can be concluded that submicroscopic bubbles (or "holes") are created and disappear continuously in every liquid as a result of statistical thermal fluctuations. The number and sizes of such bubbles increase rapidly with temperature and especially in case the liquid becomes superheated. If the temperature and superheat are high enough the liquid may be unstable because of the increased probability for a submicroscopic bubble to surpass its critical size and spontaneous boiling may occur. Therefore, it seems plausible that a direct heating of localized regions in a liquid may produce a similar effect. Some heating of limited zones should occur as a result of the passage of an ionizing particle through the liquid.

The energy required to form a bubble in a liquid is strongly dependent on the way in which it is formed, the least energy is spent when the formation takes place isothermally and reversibly. A more plausible assumption is, however, that under the circumstances considered here, a bubble is created in an adiabatic and irreversible way [8]. The minimum energy necessary can, as will be shown later in this paper, be estimated at least roughly. For normal operating conditions of organic liquids this minimum energy is found to be in the range 100 eV–400 eV.

If one accepts that the bubbles are produced by δ-rays, one finds by using the same argument as in the discussion of the electrostatic theory that it does not seem impossible to get the energy loss of the δ-rays highly concentrated in many regions of the liquid. As far as can be concluded from experimental data, the number of bubbles along a track is considerably smaller than the number of δ-rays with energy large enough to produce bubbles. This fact can be interpreted in different ways. Firstly, all the energy may be localized enough but only part of it available within the time the bubble requires for growth to the interesting size. Secondly, the energy given up by the slowing down electrons may be distributed in such a way that only in some cases this energy becomes highly localized. Thirdly, the energy necessary for bubble production may be unexpectedly large for reasons given in a later section of this paper. It is also possible that all the effects mentioned here are of importance.

The heating of a small volume may come partly from the recombination energy of produced ions, partly from the energy release in de-exciting atoms and molecules. An experiment performed with a DC electric field across a propane bubble chamber by Goldburg [7] suggests either that the energy of recombination is of negligible importance for the production of bubbles or that the recombination takes place in a time shorter than that required for the bubbles to reach their critical sizes. The major part of the energy loss of a δ-ray in a liquid or a solid is spent for molecular or atomic excitation and as a rule this excitation energy is readily converted into kinetic energy of adjacent molecules. Therefore, the number of heated zones, "thermal spikes", should be sufficiently large for the nucleation to occur with a frequency consistent with that observed. This holds even if the time constant for ion recombination should be so large that the recombination energy cannot contribute to bubble formation. The estimates of this time constant are, according to Goldburg, crude enough to allow it to be as large as 10^{-7} sec.

The de-exciting of molecules or atoms, which can be expected to occur with a small time delay in most liquids, is more difficult in xenon and a satisfactory explanation of its behaviour is given by the special properties characteristic of monatomic liquids. Therefore, one might imagine liquid helium to behave in a similar manner. However, helium seems sensitive to ionizing radiation just as an "ordinary" liquid.

One complicating possibility, which cannot be excluded for the moment, is that bubbles may predominantly come from such domains in the liquid which as a result of spontaneous temperature fluctuations already have a temperature higher than the ambient. The probability for a δ-ray to stop in such a domain should, however, not be very large. When a bubble chamber is operated at superheats giving relatively low bubble densities the effect should hardly be noticeable because of the large energies then required in the thermal spikes.

Finally, it may happen that both the processes described above contribute to the formation of bubbles and the importance of the two processes may also be different in different liquids or at different operating conditions.

Object of the present investigation

Even if the experimental observations for the present seem to be more in favour of the thermal than the electrostatic theory of the bubble chamber, the latter theory cannot yet be excluded with certainty. The bubble initiating process is in its details unknown enough to allow for unexpected effects. It would, therefore, be of interest if one could find an experiment independent of those related in the review above which supported one or the other theory. The problem is a difficult one taking into account, for instance, the extremely short time required for the growth of a bubble to its critical size and the fact that the demonstration of the presence of ions which is so easily done in the cloud chamber seems impossible in the bubble chamber.

Experimental method

The investigation described here uses a method which was originally proposed by Glaser [4]. It depends on measuring the temperature dependence of the conditions for bubble formation. Clearly this should be feasible if it could be assumed that the number of charges produced in clusters along the path of a particle in a bubble chamber according to the electrostatic theory was roughly independent of temperature and density of the liquid or—in case the nucleation was due to "heat pulses", or thermal spikes, as required by the thermal theory—the available energy in these pulses was also roughly independent of changes in density and temperature of the liquid.

Since such assumptions evidently cannot hold strictly, it will be assumed in the following that a considerably better description of the actual process can be obtained by introducing a few correction factors. The number of charges on the bubbles should, for instance, be calculable at one density of the liquid if known at another density and the same should also be true of the energies in the termal spikes. Of course, this will be possible only for limited ranges of density and temperature.

The advantage of the method is then, as will be shown later, that it is rather independent of quantities which cannot be measured, for instance the number of the charges on the bubbles and the energy in the thermal spikes. Neither is it necessary to know accurately the surface tension of the liquid, nor the latent heat of vapourization, nor the dielectric constant. Only the changes in these properties must be known and these changes can be calculated rather accurately.

The procedure in the following will be to derive formulas from the two theories, to estimate correction factors, to calculate certain properties of the liquids when they cannot be obtained directly from tables, and finally, to bring the calculated temperature dependences in a form suitable for comparison with experiments.

Calculations of the temperature dependence

Electrostatic theory

Let us consider a bubble in a superheated liquid assuming that it has n electrical charges of the same sign on its surface, where $n > 2$, or $n = 0$. For the sake of simplicity it will be assumed that the charges are continuously and uniformly distributed over the surface of the bubble although at the low number of charges here—of the order of 6 as stated before—the charges should, if more precise calculations could be made, be treated as point charges. Here, the aim is to investigate how the bubble behaves at or near its equilibrium state, and especially in the vicinity of its critical size. The radius at the critical size is roughly 5×10^{-7} cm in most liquids under the conditions of temperature and superheat which are of interest. Under the assumptions of uniform charge distribution the bubble will be spherical in shape. Even if the charges were not smeared out evenly on the surface the deviations from the spherical form, if not too large, would probably not alter significantly the actual behaviour from that calculated on such an assumption. Arguments for this statement will be given in the discussion at the end of this paper.

The following pressures are found to act on the surface of the bubble:

(*a*) The vapour pressure, p_r, of the gas inside the bubble. This pressure is a function of the temperature of the vapour inside the bubble and the radius of curvature, r, of the surface.

(*b*) The hydrostatic pressure in the liquid, p.

(*c*) The pressure from the surface tension, $2\sigma/r$. The surface tension, σ, is assumed to be a function of temperature only and independent of the radius of curvature. At very small radii the surface tension may be a function of radius [11]. For reasons given later, the final results of the calculations will probably not be affected at all by neglecting a small dependence of the surface tension on the radius of curvature.

(*d*) The effective electrostatic pressure

$$\frac{n^2\,(1-\varepsilon/n)\,e^2}{8\,\pi\,\varepsilon\,r^4},$$

where ε = dielectric constant of the liquid, and e = charge of the electron.

For a bubble which is in static equilibrium with its surroundings in the liquid, the following relation must hold

$$(p_r-p)=(p_r-p)_n=\frac{2\,\sigma}{r}-\frac{n^2\,(1-\varepsilon/n)\,e^2}{8\,\pi\,\varepsilon\,r^4}, \tag{1}$$

In the case of no charges on the bubble this reduces to

$$(p_r-p)=\frac{2\,\sigma}{r}. \tag{2}$$

The limiting condition on the degree of superheat expressed in terms of pressure, $(p_r-p)_n$, for a bubble to grow to visible size is that

$$(p_r-p)_n=\left[\frac{27\,\pi}{2\,e^2}\right]^{\frac{1}{3}}\sigma^{\frac{4}{3}}\,\varepsilon^{\frac{1}{3}}\left[n^2\left(1-\frac{\varepsilon}{n}\right)\right]^{-\frac{1}{3}}. \tag{3}$$

The condition given by (3) is schematically illustrated in Fig. 1. For a given number of charges, n, a stability curve as shown in the figure is obtained. There exists a lower limit for the degree of superheat, $(p_r-p)_n$, which must be exceeded if a bubble is not to collapse in the liquid. If one assumes an isothermal formation, a bubble at the superheat corresponding to point A in the figure can always grow to visible size regardless of its initial size, whereas a bubble at the superheat B will grow only to a certain radius determined by the stability curve, and will remain stable at that radius as long as the charges on its surface are not removed. This behaviour will be of some importance for the conclusions drawn in a following paragraph.

The critical radius of a charged bubble, r_1, is given by the expression,

$$r_1=\left[\frac{n^2\,(1-\varepsilon/n)\,e^2}{4\,\pi\,\varepsilon\,\sigma}\right]^{\frac{1}{3}}=\frac{3\,\sigma}{2\,(p_r-p)_n}, \tag{4}$$

and denoting the critical radius of an uncharged bubble by r_2,

$$r_2=\frac{2\,\sigma}{(p_r-p)}. \tag{5}$$

Even if (3) could be expected to hold rigorously a word of caution must be inserted before it is used to find the variation of $(p_r-p)_n$ with temperature. It might, in fact, happen that in a growing bubble the temperature of the surface and the vapour inside the bubble could have changed from the inititial values during the time required for the bubble to reach its critical size since the growth process consumes energy. It is

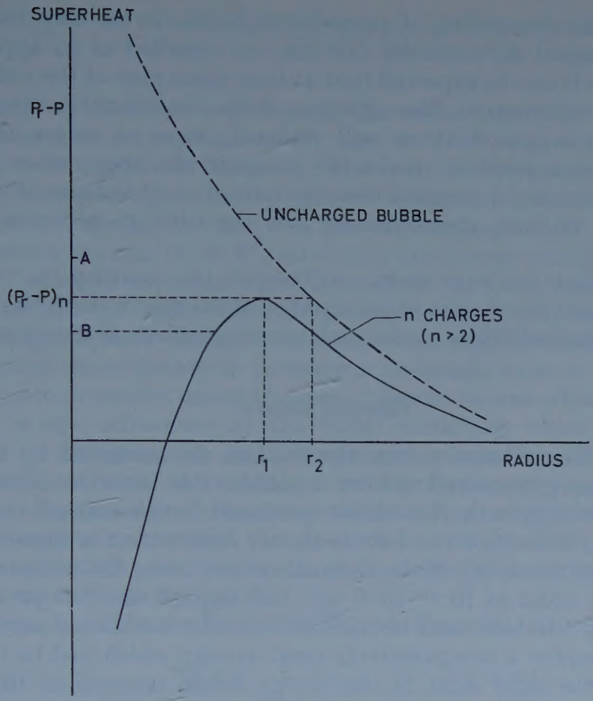

Fig. 1. Stability curves for bubbles in a superheated liquid as a function of their radii. The superheat is expressed in terms of the pressure. A bubble with n charges on its surface will at the superheat valid at point A grow to visible size, regardless of its initial radius while a bubble at superheat B will grow to a definite radius, at which it is stable until the charges on its surface have disappeared. The critical radius of a charged bubble is denoted by r_1, that of an uncharged by r_2.

difficult to estimate the electrostatic energy of a bubble because the initial and final charge configurations are unknown as well as the radius from which the bubble starts, the knowledge of which would be sufficient when uniform charge distribution is assumed. Anyhow, one must allow for the possibility that the electrostatic energy cannot supply more than some fraction of the energy of bubble creation and, therefore, it cannot be excluded that some additional energy in the form of heat has to be taken from the surrounding liquid. If this is to occur there must at some stage of the bubble growth exist a temperature gradient outside the surface of the bubble. A charged bubble, which by an isothermal process could just pass the critical radius under limiting conditions on the superheat, would in case it is subject to a temperature drop grow to a radius somewhat smaller than the critical and find itself in a situation where during the first moment it could only reach a radius which is determined by the stability curve and which is somewhat smaller than the critical. Thus, such a bubble would behave like the bubble at superheat B in Fig. 1 as discussed earlier. If now the charges on the bubble surface remain for the time necessary for the temperature gradient outside the surface to vanish, the growth to a larger radius than the critical is possible. Another reason why the temperature drop in the bubble, if occurring at all, should be small is that some heat can be expected to be supplied to the bubble by the de-exciting of atoms or molecules in the way discussed earlier. Of course, this assumed development can only be regarded as an approximation of the actual one since it can be expected that at least some part of the initial net charge will be lost by recombination. Nevertheless, if the electrostatic theory is correct, effects of the type described above will evidently tend to reduce the effect of a temperature drop and probably make the temperature drop rather small. In the following it will therefore be assumed that the formation of bubbles at the lower limit of superheat can, at least approximately, be regarded as occurring at constant temperature.

The expression (3) is not very useful yet because the quantity $(p_r - p)_n$ cannot be measured directly and the change of the number of charges, n, with the density of the liquid has to be estimated. First, the formulas from the thermal theory will be derived.

Thermal theory

On the basis of the assumption that the bubbles are nucleated by thermal spikes in the liquid, the energy required to form a bubble of at least the critical size can be estimated. The rate of growth of a bubble produced in this manner must, according to Seitz [8], be very fast since the bubble should form within a time which is of the order of the time for the decay of the thermal spikes. Since the estimates of the time constant are of the order of 10^{-10}–10^{-11} sec, this implies that the process should be treated as a closely adiabatic and irreversible one. An isothermal, reversible bubble formation would require a comparatively small energy which had to be supplied by the ionizing particles since most of the energy would come from the surrounding liquid.

That energy in a thermal spike which is of interest here is, of course, only that part of the energy in the spike which is available within the time necessary for a bubble to reach its critical size. The following analysis will be based on the assumption that the whole available energy in a thermal spike has been consumed during the interesting stage of the bubble growth.

The minimum energy, W_m, required to form a bubble of the critical radius r_2, at the absolute temperature T in the liquid, and in agreement with the adiabatic growth assumed above can be written

$$W_m = W_1 + W_2 + W_3 (+ W_4), \tag{6}$$

where W_1 = energy of the surface

$$= 4\pi r_2^2 \left(\sigma - T \frac{d\sigma}{dT}\right), \tag{7 a}$$

W_2 = energy required to vaporize the gas inside the bubble

$$= \frac{4\pi r_2^3}{3} L \varrho_v', \tag{7 b}$$

where L = latent heat of vaporization,

ϱ_v' = density of the vapour inside the bubble,

W_3 = net work done against the hydrostatic pressure

$$= \frac{4\pi r_2^3}{3}(p - p_r), \tag{7 c}$$

and W_4 = energy which must be present in the initial heat pulse because the liquid may not be treated as incompressible. W_4 may be quite large when the influence of viscosity cannot be neglected. However, that part of the energy W_4 which is not converted into heat in a region, nearly equal to the critical volume of the bubble, will be absorbed somewhere farther away in the surrounding liquid. Consequently, W_4 should not enter in the sum (6) if W_m is to mean the energy consumed by the bubble itself. If W_4 is included the meaning of W_m will be the total energy which must be available in the thermal spike. According to Seitz [8] it can be expected that in propane the energy W_4 is not negligible in comparison with the other energies although he points out that the influence of viscosity is uncertain because no accurate values of the viscosity are available, under the special circumstances which one obtains here. Probably, W_4 is also influenced by the initial conditions which must be specified in calculations for the primarily heated zone giving a bubble. If W_4 should be too high, say, 4 keV or more, the model of bubble formation considered here could hardly work at all since as a rule there are too few δ-rays along the path of a particle with enough energy to account for the observed bubble densities. In such a case the process had to be treated quite differently since most of the energy for bubble formation had to be taken from the liquid.

In the following it will be assumed that $W_4 = 0$. From (5), (6) and (7a–c) it is found

$$W_m = \frac{32\pi\sigma^3}{3(p_r - p)^2}\left[\frac{L\varrho_v'}{(p_r - p)} + \frac{1}{2} - \frac{3}{2}\frac{T}{\sigma}\frac{d\sigma}{dT}\right]. \tag{8}$$

This is an equation of the third degree in $(p_r - p)$ and can with $(p_r - p) = \Delta p_r$ be written in the form

$$\Delta p_r^3 - a(T, W_m)\Delta p_r - b(T, W_m) = 0, \tag{9}$$

where
$$a(T, W_m) = \frac{32\pi\sigma^3}{3W_m}\left(\frac{1}{2} - \frac{3}{2}\frac{T}{\sigma}\frac{d\sigma}{dT}\right)$$

and
$$b(T, W_m) = \frac{32\pi\sigma^3}{3W_m}L\varrho_v'.$$

Before proceeding any further with (9) it can be noted that analogous to $(p_r - p)_n$ in the electrostatic case, Δp_r cannot be measured directly and the influence of the change of density of the liquid on the energy in the thermal spikes should be taken into account. Therefore such corrections will be treated first.

Correction on pressure drops

The saturation pressure of the vapour inside a bubble, p_r, is somewhat lower than that over a plane surface, p_∞. The quantity experimentally determined is $(p_\infty - p)$ and therefore a relation between $(p_r - p)$ and $(p_\infty - p)$ must be established. The expression for p_r generally used in the theory of the cloud chamber [12] is inadequate here for two reasons. Firstly, the gas inside a bubble cannot be treated as ideal because it is assumed to be at its saturation pressure; secondly, the density of the vapour is not negligible in comparison with that of the liquid. A better approximation can be obtained from the following expression [13], valid for a charged bubble.

$$(p_r)_n = p_\infty - \left[\frac{2\,\sigma}{r} - \frac{n^2\left(1-\frac{\varepsilon}{n}\right)e^2}{8\,\pi\,\varepsilon\,r^4}\right]\cdot\frac{\varrho_v'}{\varrho_l - \varrho_v'}, \tag{10 a}$$

where ϱ_l = density of the liquid.

ϱ_v' is somewhat lower than ϱ_v, the density of saturated vapour at the pressure p_∞. Since it is difficult to estimate ϱ_v' and ϱ_v is known accurately it will in the following be assumed that

$$\frac{\varrho_v'}{\varrho_l - \varrho_v'} \approx \frac{\varrho_v}{\varrho_l} \tag{10 b}$$

and that in (7 b) ϱ_v can be used instead of ϱ_v'. With this modification of (10 a) it follows from (1) and (10 a) that

$$(p_r - p)_n = (p_r - p) = (p_\infty - p)\left(1 + \frac{\varrho_v}{\varrho_l}\right)^{-1}. \tag{11}$$

The correction to be applied to the pressure difference between the pressure inside the bubble and the hydrostatic pressure is thus the same for a charged as for an uncharged bubble at the limiting conditions (1) or (2) and at the corresponding critical radii given by (4) or (5) since the right-hand side of (11) does not contain n, the number of electrical charges.

Correction for variation of density of the liquid

Of interest for the present investigation is the number of domains in the liquid along the path of a particle which fulfill at least the condition for production of a bubble. An excess of either charges on the primary bubbles or heat in the thermal spikes will not be of any use since not more than one bubble can arise from each domain. The method used for comparing the experimental and calculated temperature dependences would be quite straightforward and simple provided that the density were the same at all pressures and temperatures for a liquid in the region investigated. Then with sufficient accuracy the number of charges on the bubbles could be regarded as independent of temperature; and, approximately, the energies in the thermal spikes would also be independent of temperature. The temperature dependence of the operating conditions could, in such a case, be easily found. One immediately finds when analysing results on sensitivity measurements in a bubble chamber that it is difficult to determine accurately the lowest pressure drop which is required to make the chamber sensitive to ionizing radiation. If now the density of the liquid were the same at the different temperatures investigated one would just have to compare the pressure drops required to obtain any given, constant bubble density.

Unfortunately, the density of the liquid will be affected by a change of temperature and hydrostatic pressure consistent with suitable operating conditions of the bubble chamber. Therefore, the following assumptions will be made.

1. The number of ion clusters or thermal spikes per unit length along the path of an ionizing particle is proportional to the density of the liquid but otherwise independent of temperature.

2. The number of charges in the ion clusters or the available energy in the thermal spikes is proportional to the density of the liquid but otherwise independent of temperature.

The justification for assumption 1 is that it is consistent with the δ-ray density produced by a charged particle, and for assumption 2 that the energy loss of a charged particle is proportional to the density of the medium traversed. Within a given volume of a liquid one should, therefore, in agreement with assumption 2, expect the number of charges with the same sign to be given approximately by

$$n = \frac{n_0 \varrho_l}{\varrho_0} \tag{12 a}$$

and, correspondingly, the energy in the thermal spikes by

$$W = \frac{W_0 \varrho_l}{\varrho_0}, \tag{12 b}$$

where n_0, W_0 and ϱ_0 are constants with the meaning of the number of charges on a bubble, the energy in a thermal spike, and the density of the liquid (neglecting the influence of pressure), all referred to an arbitrary temperature T_0.

From assumption 1 it follows that a comparison of measured pressure drops at different temperatures should be made not at constant bubble density but at the bubble density $g = g_0\,(\varrho_l/\varrho_0)$, where g_0 is some bubble density chosen at temperature T_0, and from assumption 2 that in equation (3) n should be replaced with (12a), and in (9) W_m with (12b) (replacing W with W_m and W_0 with W_{0m} respectively).

Properties of liquids used

The investigation was performed with the following three liquids, propane (C_3H_8), Freon-12 (CCl_2F_2) and sulphurhexafluoride (SF_6). The reasons for choosing these liquids were that the bubble chamber available could only be used at temperatures not too far from room temperature and that these liquids could easily be obtained with good purity. Experimental details not quoted here or in the following can be found in a paper by Alfredsson and Johansson [14].

Some of the values of the properties of the liquids which are required for the numerical calculations can be obtained directly from standard tables [15, 16] but care must be taken that only such measurements are accepted which appear accurate.

For the sake of simplicity the influence of pressure on the density, the surface tension, and the dielectric constant of the liquid will be neglected in the following. These properties of the liquids are not strongly affected by moderate changes of pressure. Furthermore, the changes in these properties can be expected to be about the same at the different temperatures at which the comparison of pressure drops will be made.

No reliable measurements on the temperature dependence of the surface tension of the liquids used here seem to exist in the temperature ranges of interest. The variation of the surface tension with temperature was, therefore, obtained by indirect methods.

Over a large range of temperature the surface tension of a normal, that is non-associated liquid, can be approximated by

$$\sigma = \sigma_0 \left(1 - \frac{T}{T_c}\right)^{\gamma} = s_0 \, (T_c - T)^{\gamma}, \tag{13}$$

where σ_0, s_0, and γ are constants, T the temperature of the liquid, and T_c its critical temperature, both referred to the absolute temperature scale.

The surface tension was calculated by using the well-known densities of the liquid and the vapour by the formulas [13]

$$\sigma = \frac{k_E \, (T_c - 5 - T)}{\left[\frac{M}{\varrho_l}\right]^{\frac{2}{3}}} \tag{14 a}$$

and

$$\sigma^{\frac{1}{4}} = \frac{[P] \, (\varrho_l - \varrho_v)}{M}, \tag{14 b}$$

where k_E = Eötvös constant = 2.1 ergs deg^{-1} $\text{mole}^{-\frac{2}{3}}$,
M = molecular weight of the liquid,
$[P]$ = constant = the parachor for the liquid.

k_E and $[P]$ are not known accurately (and are probably slightly dependent of temperature) but seem accurate enough since in the first place only the change of surface tension with temperature is sought.

The values of γ in (13) obtained from (14a) or (14b) agreed for the liquids considered here within less than 2 %, and the mean values were accepted. γ is quoted in Table 1 together with the surface tension at the temperature T_0.

The latent heat of vaporization, L, was calculated from the Clausius–Clapeyron equation and the dielectric constant, ε, from the Clasius–Mosotti formula for the molecular polarizability [15]. For comparison the values of L and ε at the chosen temperature, T_0, are shown in Table 1.

Table 1.

Liquid	T_0 °K	T_c °K	$\sigma(T_0)$ dynes/cm	γ	$L(T_0)$ ergs/mole	$\varepsilon(T_0)$
Propane	332.5	368.8	3.07	1.24	12.06 10^{10}	1.48
Freon-12	345.5	384.7	3.07	1.27	12.09 10^{10}	1.85
Sulphurhexafluoride	285.5	318.7	2.75	1.27	10.82 10^{10}	1.58

Estimate of W_{0m}

The coefficient a and b in (9) are dependent on the bubble formation energy, W_m, assumed. W_{0m} was therefore calculated from the experimentally obtained value of $\Delta p = (p_\infty - p)$ at the bubble density 10 bubbles cm and at the temperature T_0, as given in the preceding sections with the use of equations (9) and (11). Fortunately, in the representation of the temperature dependence which will be employed, the value of W_{0m} is not very critical, as will be made clear later.

Temperature change of liquid during expansion

A complicating fact which must be treated in some detail is that the production of a superheated state by a rapid expansion of the liquid will cause a temperature change. The temperature of the liquid is thus not the same at the moment when the ionizing particles are sent through it as it was before expansion.

If the expansion were carried out adiabatically and reversibly the temperature change could easily be found [17] from

$$\Delta T = \frac{T\,\bar{v}_l\,\bar{\delta}}{\bar{c}_p}(p_f - p_i), \tag{15}$$

where v_l = molar volume of the liquid,

$$\delta = \frac{1}{v_l}\left(\frac{\partial v_l}{\partial T}\right)_p,$$

c_p = molar heat capacity of the liquid at constant pressure.

A bar over a letter denotes the average value. p_i and p_f are the initial and final pressures respectively. The lack of measured values of δ for the liquids in question, which could be considered as only approximately correct in the superheated, unstable state combined with the fact that the expansion, although closely adiabatic, is not a reversible one, will make results obtained from (15) rather uncertain. From P.V.T. data available on iso-pentane [16] a rough estimate of δ for propane can be found if the two liquids are compared at the same reduced temperature T/T_c. The temperature change for a pressure drop of 15 atm in propane at 60°C should, estimated by (15) be of the order of -1.5°K.

In a bubble chamber there are complicating factors present (e.g. losses due to internal friction in the liquid, work done on the liquid during compression) which to some extent are dependent on the operating conditions of a special chamber. Therefore, the temperature change was determined experimentally for propane by measuring the change in hydrostatic pressure which is required to obtain a given bubble density when the preexpansion pressure was varied. It was done at 60°C pre-expansion temperature, and the result found was

$$\frac{d(\Delta T)}{d\,p_i} = -(0.047 \pm 0.015)^\circ\mathrm{K/atm}.$$

When the chamber is expanded from $p_i = p_\infty + 5$ atm to $p_f = 10$ atm, this implies a temperature drop of (0.7 ± 0.2)°K when linear extrapolation is used. In the temperature region of interest $\bar{\delta}$ in (15) increases with temperature for the liquids here but since $|p_f - p_i|$ decreases with temperature so that $\bar{\delta}\cdot|p_f - p_i|$ should be nearly constant it will be assumed that the temperature drop is the same in the whole temperature region investigated in propane. For the sake of simplicity it will also be assumed that the temperature drop is approximately the same for all the three liquids investigated here because they can be considered to be in what is in thermodynamics called corresponding states.

Expression for the temperature dependence

To solve (9) which is obtained from the thermal theory one has to assign definite temperatures to the surface of a bubble and the vapour inside it. In the discussion preceding the deriving of (9), it was assumed that the energy in a thermal spike was just enough to create a bubble of critical size. In order to simplify the calculations it will now be assumed that the temperatures in the liquid, at the surface of a bubble, and in the vapour inside it are all the same. From (9), (11), (12b), and (13) and with a fixed value of W_{0m} as explained earlier, as well as from (3), where $(1-[\varepsilon/n])$ is treated as a constant, (11), (12a), and (13) it is found that both the thermal and the electrostatic theories predict temperature dependences of the pressure drop, which with good accuracy can be written in the convenient form

$$\log_{10}\Delta p = \text{const} + K_i \log_{10}(T_c - T), \tag{16}$$

where $\Delta p = (p_\infty - p)$ and $i = 1, 2$.

K_1 denotes the slope as found from the electrostatic and K_2 that from the thermal theory. Calculated values of K_i are given in Table 2.

It may be noted here that K_2 is slightly influenced by the choice of the energy W_{0m} which will appear in (9). It is therefore, in contrast to K_1, also dependent on the constant σ_0 (or s_0) in (13). However, a change of W_{0m} by a factor of 2 (or the same change in σ^3) alters K_2 with only about 2 %, which is hardly experimentally detectable, and hence it is not necessary to know accurate values of neither energies for bubble production nor the surface tension.

Comparison of calculations with experiments

The relations between the bubble density and pressure drops at different temperatures obtained in propane, Freon-12 and sulphurhexafluoride [14] were used for the comparison of calculations with experiments. It was considered desirable to investigate the behaviour of one of the liquids more thoroughly, and therefore, the measurements in propane originally performed at the (pre-expansion) temperatures 52, 56, 60, 64, and 68°C were extended to comprise also to the temperatures 54, 58, 62, and 66°C. When a bubble density $\leqslant 10$ bubbles/cm is used for the comparison of pressure drops the large number of measured points gives in total 9 points for the determination of the temperature dependence for this liquid.

Fig. 2 shows the bubble density, g, in propane as a function of the measured pressure drop, Δp, with the estimated temperature, T, of the expanded liquid as a parameter. In the figure is also indicated how the pressure drops to be compared at the different temperatures are obtained. This is illustrated at the bubble density which corresponds to 10 bubbles/cm at $T_0 (= 59.3°\text{C})$ in agreement with assumption 1 in the section on the correction for the change of density of the liquid.

The uncertainty in the pressure measurements for the individual points in Fig. 2 is estimated to about ± 0.10 atm (standard deviation) corresponding to about 0.7 % of the total measured pressure change [14]. It should also be noted that along an "isotherm" the errors in the measurements of pressure drops are not uncorrelated.

In Fig. 3 $\log_{10}\Delta p$ has been plotted versus $\log_{10}(T_c - T)$ for the pressure drops obtained for different bubble densities in propane. The measured values of Δp for a given bubble density can, considering that the standard deviation of the measure-

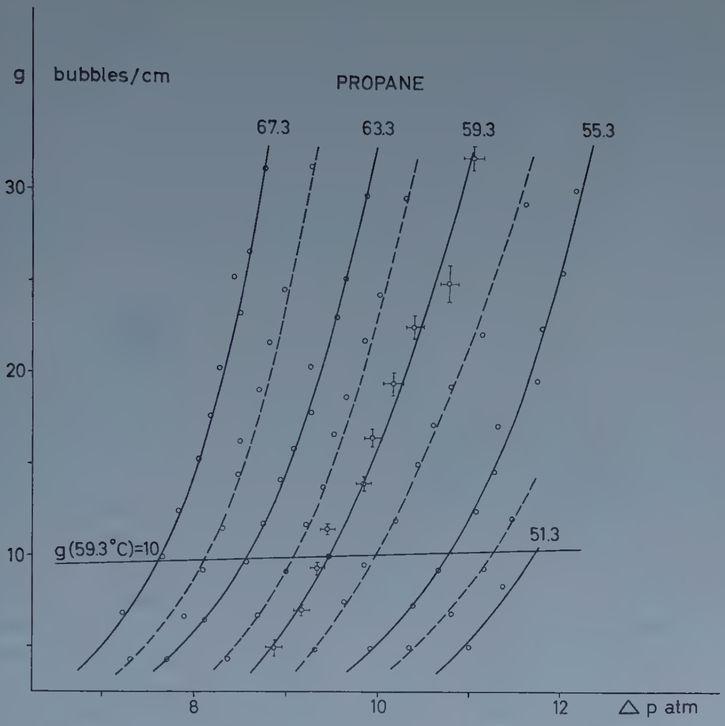

Fig. 2. Bubble density curves in propane as a function of the pressure drop, Δp, with the estimated temperature of the expanded liquid, T (in °C), as a parameter. The figure shows an example of how the pressure drops to be compared are obtained. The start is made from the bubble density 10 bubbles/cm at the temperature T_0 ($=59.3$°C). The pressure drops sought at a curve corresponding to temperature T is found at the bubble density 10 ϱ_l/ϱ_0 bubbles/cm as explained in the text.

ments is about 0.10 atm, be approximated by straight lines. The numbers at the lines denote the bubble density at the temperature T_0. In the figure the slopes of the lines are also given. $K(g)$ denotes the slope obtained when one starts from a bubble density of g bubbles/cm at the temperature T_0. The mean value of the slopes of the different lines, $\bar{K}$, is quoted in Table 2.

The experimental results obtained on Freon-12 and sulphurhexafluoride were treated in the same way as those for propane. While for Freon-12 a comparison of pressure drops could be done at the same bubble densities as for propane it was in the case of sulphurhexafluoride necessary to use the bubble densities 16 and 25 bubbles/cm instead of 18 and 30 bubbles/cm. The bubble densities quoted are, of course, referred to the temperatures T_0 as given in Table 1. Also for these liquids the measured values of the pressure drops can be approximated with straight lines. The standard deviation of the measured values of Δp is practically the same as for propane but the number of measured temperatures lower. The slopes obtained at different bubble densities vary somewhat and as in the case of propane (Fig. 3) the slopes have their largest values at the lowest bubble densities. This variation amounts to about 10 % for Freon-12 and 12 % for sulphurhexafluoride. Since the variation seems to be

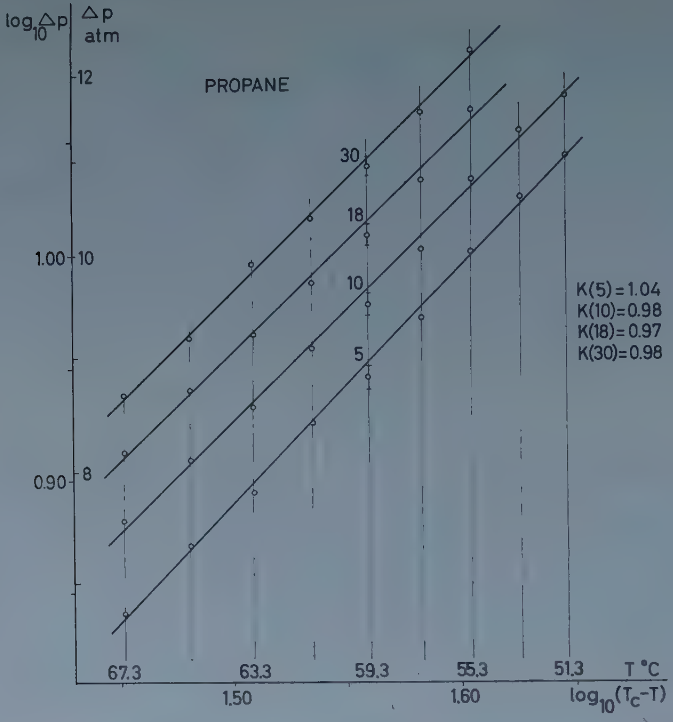

Fig. 3. A plot of $\log_{10} \Delta p$ versus $\log_{10} (T_c - T)$ for the measurements on propane and with the bubble density at $T_0 (= 59.3°\mathrm{C})$ as a parameter. The uncertainties in the pressure drop measurements have been indicated in the figure at the temperature T_0. The uncertainties are very nearly the same for all the measured points. The value of the slope of a line corresponding to bubble density g bubbles/cm at T_0 is denoted by $K(g)$, and the slopes obtained from the different lines are also given.

within the estimated error limits for the slope, one cannot with certainty draw the conclusion that it is due to a real physical effect. The mean values of the slopes for these two liquids are also given in Table 2.

The limits of error quoted in Table 2 can only be regarded as approximate because of the large number of quantities implicitly involved in the calculations of the slopes. The errors of these quantities are also to some extent correlated.

Table 2.

Liquid	Calculated values		Experimental values
	K_1	K_2	$\overline{K}$
Propane	1.44	1.00	0.99±0.10
Freon-12	1.52	1.06	1.10±0.12
Sulphurhexafluoride	1.46	0.98	0.90±0.13

As can be seen from Table 2 the agreement is striking between the temperature dependence as calculated from the thermal theory and that obtained experimentally. It can be noted here that if the process of bubble formation in the thermal case had been considered to take place isothermally and reversibly, instead of adiabatically and irreversibly, the calculated value of the slope K in (16) would have been about 1.65 for propane and nearly the same for the other liquids. Since this value is quite far from the experimental one in propane, and the same is true of the other liquids one can with certainty conclude that the bubble formation cannot even roughly be described as an isothermal and reversible one.

Discussion

The values of the slope, K, calculated from (16) are of course appropriate only when the assumptions hold which have been made to obtain the equations used in (16). It is then natural to investigate what modifications of the assumptions, concerning the electrostatic theory, are necessary to bring the calculated values in agreement with the experimental ones and also how uncertainties in some of the calculated properties of the liquids used influence the results of the calculations.

Firstly, it should be pointed out that the slope K in (16) remains practically unchanged even when larger temperature intervals than here are considered for the liquids. The experimental values of the slope are, however, dependent on the temperature assumed in the bubbles mainly because of the rapid temperature variation of the saturation pressure, p_∞, which affects the calculated pressure drop, $\Delta p = (p_\infty - p)$.

Secondly, the percentage error of γ in (13) influences the calculated value of the slope, K, with practically the same amount. This error can hardly amount to more than a few per cent.

Thirdly, formula (3) which gives the temperature dependence according to the electrostatic theory contains n, the number of charges on the bubbles. Since n must be a whole number one may think that this would lead to discontinuities in the bubble density curves in Fig. 2. The absence of detectable discontinuities can probably not be taken too seriously as an argument against the electrostatic theory even if (3) were correct. The actual behaviour of the bubbles is influenced by statistical fluctuations in the temperature of the bubble, in the number of molecules inside it, etc.

The discrepancy between the experimentally found temperature dependence and that calculated from the electrostatic theory may, in case this theory were correct, be due to:

(*a*) The temperature of the surface of the bubble and the vapour inside it might at the critical size of the bubble be lower than that of the liquid. With the use of the experimentally found temperature change due to the expansion of the liquid (−0.7°K) it is then necessary to assume that the temperature of the bubble must be about 4°K lower than that of the liquid. This seems a rather large value considering that the temperature drop should be confined to a distance of the order of the critical radius resulting in a very high temperature gradient.

(*b*) The number of charges on the bubbles could decrease more rapidly with the density of the liquid than given by (11a). However, if such an effect was alone responsible for the observed discrepancy, the decrease of the number of charges with the density must be several times more rapid than that assumed, which is difficult to explain. If recombination effects are of importance this rapid variation should be at least partially cancelled by the correspondingly larger time constant for the recombination because of the lower ion concentration in the ion clusters at the higher temperatures.

(*c*) The experimentally determined temperature drop could be too low. As for (*a*), it is found that if this alone was responsible for the discrepancy the temperature change must in total amount to $-4.7°K$, which can be regarded as impossible.

(*d*) The bubbles are not spherical in shape. If the deformation is not very large at the critical size, the effect caused by the deformation should be small. The variation of the number of charges on the surfaces of the bubbles with density of the liquid is, when estimated from (12a), not large ($\simeq 7\%$) in the temperature region of interest. The critical radii calculated by (4) do neither vary very much ($\simeq 10\%$) under the conditions at which the pressure drops are compared. As the bubbles can be expected to carry roughly the same number of charges and have practically the same radii they should be of about the same shape, which is sufficient for the calculations here to be valid.

(*e*) The combined effect of disturbances of types (*a*)–(*d*). Unfortunately, it seems in such a case impossible to estimate the probability for a result as that obtained experimentally.

It can also be noted that in case the surface tension of a bubble were radius dependent that would hardly be noticeable in this investigation. The main effect would be that all values of the surface tension would change by practically the same amount which for reasons given in connection with eq. (16) should not influence calculated values of the slope very much.

The assumption, which was made before the numerical solution of (9), that the temperature of a bubble of critical size is equal to that of the liquid, is somewhat arbitrary. Actually, there may be a small temperature difference. The description of the bubble formation in the way assumed is, however, supported by the results of the experiments.

Conclusion

The results obtained in this investigation suggest that the thermal theory of the bubble chamber gives a description of the bubble nucleating, which is satisfactory. Although some contribution to the bubble formation from an electrostatic process of the kind usually assumed cannot be entirely excluded, the conclusion to be drawn is that most bubbles are formed by the action of thermal spikes in the liquid. Another fact which could possibly also be taken as an indication of the correctness of the thermal theory is the simple formula found for the bubble density in another investigation [14]. The formula gives the bubble density in terms of the minimum energy required for bubble production when the energy is calculated according to the conditions valid by a thermal nucleation.

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